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Abstract: A multi-season field trial was carried out to investigate the effect of the amendment of biochar, lime, ash and washed biochar on the growth of maize. A degraded, strongly acidic Ultisol (pHKCl 3.60), with a relatively high exchangeable aluminium content (2.4 cmolc/kg) and a low exchangeable calcium content (0.99 cmolc/kg), was used. Soil was treated once at the beginning of the field trial and crop growth was monitored over seven planting seasons (PS). All treatments increased maize yield. The average increases were; seven times for biochar, five times for lime, five times for washed biochar and eight times for ash treatment, when compared to the control across all PS. The effect of biochar, lime and ash treatments on maize yield were sustained over the seven PS. Soil pHKCl was significantly increased ($p < 0.05$ level) following the addition of all of the amendment materials. All treatments significantly reduced the concentration of Al^{3+} when compared to the control ($p < 0.05$), with the lowest concentrations for the lime and ash treatments. The ash treatment also increased the concentration of macronutrients (K, P and Mg) to the greatest extent. Results showed that there was a clear liming effect at play. The better performance of biochar compared to lime, despite lime having the highest pH and the lowest Al^{3+} concentration, can be explained by the additional K, Mg and P the biochar adds to the soil. Results also showed a clear nutrient addition effect where ash added the most nutrients. Overall, this work supports the fact that small scale farmers in Indonesia should produce biochar from their waste agricultural materials. Doing so not only provides an increase in crop productivity, but also sequesters carbon resulting in the best overall environmental benefit.

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The effect of biochar, lime and ash on maize yield in a long-term field trial in a Ultisol in the humid tropics

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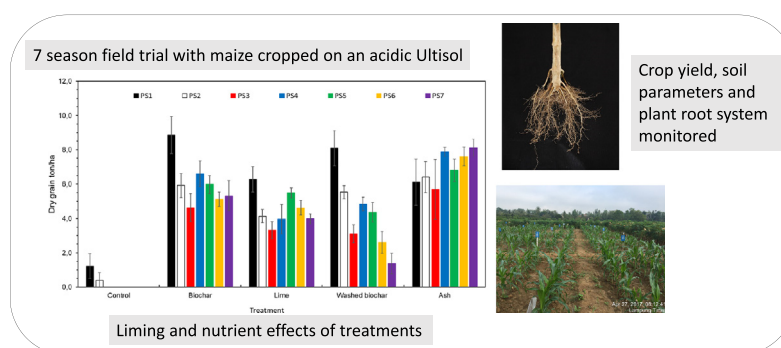
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HIGHLIGHTS

- Biochar, lime, ash and washed biochar were added to an acidic soil.
- Average maize yield increased 7 times for biochar, 5 times for lime and washed biochar and 8 times for ash.
- Clear liming effect of biochar with an increase in pH and decrease in available Al^{3+} .
- Clear nutrient addition effect of ash with an increase in K, P, Mg and Ca concentrations.
- Ash had the strongest effect on root angle opening, area and stem diameter.

GRAPHICAL ABSTRACT



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ABSTRACT

A multi-season field trial was carried out to investigate the effect of the amendment of biochar, lime, ash and washed biochar on the growth of maize. A degraded, strongly acidic Ultisol (pH_{KCl} 3.60), with a relatively high exchangeable aluminium content (2.4 $cmol_c/kg$) and a low exchangeable calcium content (0.99 $cmol_c/kg$), was used. Soil was treated once at the beginning of the field trial and crop growth was monitored over seven planting seasons (PS). All treatments increased maize yield. The average increases were; seven times for biochar, five times for lime, five times for washed biochar and eight times for ash treatment, when compared to the control across all PS. The effect of biochar, lime and ash treatments on maize yield were sustained over the seven PS. Soil pH_{KCl} was significantly increased ($p < 0.05$ level) following the addition of all of the amendment materials. All treatments significantly reduced the concentration of Al^{3+} when compared to the control ($p < 0.05$), with the lowest concentrations for the lime and ash treatments. The ash treatment also increased the concentration of macronutrients (K, P and Mg) to the greatest extent. Results showed that there was a clear liming effect at play. The better performance of biochar compared to lime, despite lime having the highest pH and the lowest Al^{3+} concentration, can be explained by the additional K, Mg and P the biochar adds to the soil. Results also showed a clear nutrient addition effect where ash added the most nutrients. Overall, this work supports the

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fact that small scale farmers in Indonesia should produce biochar from their waste agricultural materials. Doing so not only provides an increase in crop productivity, but also sequesters carbon resulting in the best overall environmental benefit.

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1. Introduction

Biochar is the solid carbonaceous material that remains following the pyrolysis of biomass. It has been amended to both tropical and temperate soils in order to sequester carbon as well as improving soil properties and crop yields, with varying degrees of success. Several meta-analyses focusing on the influence of biochar on soil physicochemical properties and crop yield have been published recently in order to disentangle mechanisms behind the effects (Biederman and Stanley Harpole, 2013; Crane-Droesch et al., 2013). The meta-analysis by (Jeffery et al., 2011) reported a small but positive effect on crop growth in both acidic and neutral tropical and temperate soils, with a grand mean increase of approximately 10%. A similar finding was reported by Hagemann et al. (2017) as they showed an 18% mean yield increase following the addition of biochar to diverse soils. In a study with 371 independent experiments taken from 114 published articles, reported that the addition of biochar to soils resulted in increased aboveground productivity, crop yield, nodulation, plant K and P as well as soil P, K, N and C contents (Biederman and Stanley Harpole, 2013). The authors also reported a better effect of the amendment of biochar in tropical compared to temperate areas. This spatial disparity was also highlighted by (Jeffery et al., 2017) who reported a grand mean increase in yield of 25% for tropical soils with a median biochar application of 15 t/ha. This compared to a grand mean increase of 13% for all data which was based on 1125 observations from 109 articles.

Acidic soils comprise approximately 50% of the total area of global arable land (Dai et al., 2017), however their inherent properties have been postulated to reduce crop production by between 30 and 40% (Berihun et al., 2017). Acidic soils are common in tropical areas and their low pH, high available Al^{3+} contents and limited cation exchange capacity (CEC) are the main factors that limit plant growth (Borchard et al., 2014). Free Al^{3+} ions in soils, increasing in concentration at decreasing pH, inhibit root cell expansion, elongation and division resulting in limited water and nutrient uptake (Dai et al., 2017). When added to acid soil, biochar can overcome Al toxicity, due to its liming effect, which results in an increase in soil pH and a decrease in concentration and toxicity of Al^{3+} . Free Al^{3+} concentrations decrease sharply at $\text{pH} > 4.2$ (Gruba and Mulder, 2008; Gruba et al., 2013). Both short-term and long-term liming effects have been reported where the short-term effect has been attributed to the ash or inorganic phase in the biochar and the long-term liming effect has been attributed to the presence of oxygenated functional groups (Berek and Hue, 2016). Concurrent with the reduction in Al^{3+} toxicity, the liming effect can also overcome phosphorous deficiency caused by fixation by iron and aluminium oxides in low pH soils (Yao et al., 2019). Biochar amendment also increases the negative charge on soil minerals and thus enhances the low cation exchange capacity (CEC) inherent to acidic tropical soils. In addition, biochar itself is negatively charged (6–59 cmol/kg; (Munera-Echeverri et al., 2018)) and its addition to soil contributes to increasing soil CEC. It has been postulated that the increased CEC, caused by amendment of biochar to tropical soils can enhance NH_4^+ adsorption and subsequently reduce N leaching (Borchard et al., 2014).

The biochar literature is vast, however long term (i.e. over several cropping cycles) field studies that have focused on the growth of maize are relatively scarce (Major et al., 2010b; Haefele et al., 2011; Abiven et al., 2015; Cornelissen et al., 2018; Pandit et al., 2018). A five-season field trial in Sumatra, Indonesia reported a positive effect of up to 15 t/ha biochar amendment in the second, third and fourth cropping cycles for maize. The soil used was an acidic ultisol (pH 3.6) and the

amendment of biochar was believed to alleviate soil acidity, confirmed by the positive relationship between yield and Ca/Al ratio, base saturation and exchangeable K (Cornelissen et al., 2018). Major et al., (2010) reported increased maize yields for the second, third and fourth cropping cycles when 20 t/ha biochar was added to a Colombian savanna Oxisol with a pH of 3.9. The authors attributed the positive effect to an increase in available Ca and Mg caused by the amendment of biochar. In a three-year intercropping field trial in Nepal, positive effects on maize and mustard yield were seen in the second and third cropping cycles. The soil used in the study was moderately acidic (pH of 4.6) and the authors attributed effects to improved soil properties (available P, K, pH, CEC, percent organic carbon and base saturation) (Pandit et al., 2018). Haefele et al. (2011) carried out extensive trials in the Philippines and Thailand, reporting 16–35% increases in rice yield due to an improvement in water retention and available K and P. A two-year study carried out in Kenya reported an increase in maize grain yield following the application of biochar in combination with inorganic fertilizer which lasted for up to four years (Güereña et al., 2016). In the longest running field trial to date in Kenya, biochar was shown to have a positive effect on maize and soybean yield that were intercropped and amended with biochar and fertilizers over 10 years. The response was ascribed to an increase in pH resulting from additive effects of the fertilizer and biochar amendment (Kätterer et al., 2019).

Despite these positive results, there are also some studies that report the opposite for long term field trials with maize. Sängster et al. (2017) did not observe any significant increase in maize growth in a three-year field study when a temperate German soil was tested. The authors speculated this was because the biochar amendment improved the availability of plant nutrients, but this was not a limiting factor in the soil used (Sängster et al., 2017). A three-year field trial carried out in the United Kingdom (i.e. a temperate soil) for maize cropped in season one and grass in seasons two and three, failed to observe a significant effect on maize yield when biochar was added. The authors speculated the difference in cropping depth was the reason for the better effect on grass which itself has its roots in the biochar application zone. (Jones et al., 2012).

The overall aim of the work presented here was to investigate whether, and by which mechanisms, the amendment of biochar to an acidic Indonesian soil could increase the yield of maize, building on previous work (Cornelissen et al., 2018). More specifically, the work addresses whether an alleviation of soil acidity and aluminium toxicity (liming mechanism), or an increase of nutrients (nutrient addition) could explain observed effects, as has been suggested previously in an extensive meta-analysis (Jeffery et al., 2017). A controlled field trial over seven planting seasons with five different amendments, where soil quality changes and yield effects were measured for all amendments, was carried out. The postulated biochar liming mechanism was probed by comparing the amendment of biochar to lime (in plots with the same pH) and the postulated improvement in nutrient availability was probed by comparing the amendment of washed biochar (to remove the ash component) with pure ash produced from the same amount of feedstock. To the best of our knowledge this is the longest biochar field trial to have been carried out in Indonesia on strongly acidic soils to date, which seeks to answer mechanistic questions. In addition, the work includes the valuable shovelomics tool and microscopic analysis to support data interpretations. Shovelomics is able to probe whether changes in nutrient content are reflected in plant growth (Trachsel et al., 2011). The microscopic analysis provides information about the microstructure and chemical composition of the biochar.

2. Materials and methods

2.1. Biochar and soil

The biochar was produced from cacao shell, using a simple kiln without a retort function as previously described in detail (Alling et al., 2014; Martinsen et al., 2015; Obia et al., 2016; Cornelissen et al., 2018) at temperatures between 300 and 450 °C. The physicochemical properties of the biochar, the washed biochar, lime and ash are shown in Table S1. The soil at the experimental station is classified as a Typic Kanhapludult and is a sandy clay loam. It has high levels of exchangeable aluminium ($2.4 \text{ cmol}_c \text{ kg}^{-1}$), a CEC of $9.7 \text{ cmol}_c/\text{kg}$ and a low pH (3.6 measured in KCl). The soil contains 54% sand, 22% silt and 24% clay. The chemical properties of the soil (prior to beginning the trial) are shown in Table S1.

The following methods were used to determine the physicochemical properties of the soil and biochar; soil and biochar pH was measured in a 1:2.5 v/v slurry in water using a pH meter (Orion 2 Star, Thermo Fisher Scientific, Fort Collins, CO, USA) after overnight sedimentation and shaking. Exchangeable base cations and Al were measured in the eluate of ammonium acetate at pH 7 and ammonium nitrate for biochar and soil respectively, with a flame spectrophotometer (Perkin Elmer, AAS 3300). CEC for soil was determined by percolation with 1 M NH_4OAc (pH = 7) followed by extraction with 0.1 M NaCl after washing with alcohol. CEC for biochar was measured as i) the total amount of extractable bases after saturation with NH_4Ac (the sum of extractable cations + the truly exchangeable fraction) and ii) the amount of extractable NH_4 after saturation with NH_4Ac and subsequent extraction with 1 M KCl (the truly exchangeable fraction). Total organic carbon (TOC) and total nitrogen (TN) were determined using CHN analyzer (CHN-1000, LECO, USA). The ash content in the biochar was determined using a thermogravimetric analyzer (TGA). The samples were heated to 650 °C and held for 1 h, then the temperature raised to 900 °C and held for 45 min. The combined weight loss at these two temperatures was taken as the loss on ignition (LOI) and the percentage ash (100% less LOI) is reported on a DW basis.

2.2. Field trial design

The field trial was carried out at an experimental station (owned by the Indonesian Soil Research Institute) in the Lampung district, South Sumatra, Indonesia (KP. Taman Bogo, ec. Probolinggo, Kab. Lampung Timur, GPS coordinates: 05000.406' S; 105,029.405' E). The Lampung district has high rainfall (1796 mm) and temperatures (30 °C) throughout the year. The field trial consisted of five different treatments, referred to as; control, biochar, lime, washed biochar and ash. Five plots per treatment (plots of $2.5 \text{ m} \times 4 \text{ m}$) were established in a completely randomized block design giving 25 individual plots. All treatments received NPK fertilizer (30:30:30 kg/ha mixture at 200 kg/ha) and urea (300 kg/ha) at the start of each season. Each treatment was applied just once at the start of planting season one. The soil amendments were applied by hand using shovels to incorporate them to the soil. The control treatment received only NPK fertilizer and urea. The biochar plot received 22.5 t/ha biochar in addition to fertilizer and urea. The lime plot received 8 t/ha lime as well as fertilizer and urea, in order to adjust the pH to exactly the same as the biochar amendment. This rate was determined in a previous small-scale field trial prior to starting this main trial. The washed biochar plot received the same dose as the unwashed biochar plots following washing with water three times for 24 h each, as well as fertilizer and urea. The ash plot received 9.8 t/ha of fully burned biochar (i.e. only the ashes that remained after complete combustion of the same amount of feedstock as used for the biochar) as well as fertilizer and urea. All treatments except from the lime were produced from the same mass of feedstock. When biochar is made, both a "char" and an "ash" fraction are produced. The ash produced by complete burning was expected to contain just the "ash"

fraction. As the "char" fraction of the biochar contains mineral elements (as well as C, H and O), complete burning was expected to produce an ash with a different composition than the "ash" fraction contained in the biochar.

The field trial was carried out for seven planting seasons (referred to as PS1 to PS7) that ran from; November 2016–March 2017 (PS1), March–July 2017 (PS2), July–October 2017 (PS3), November 2017–March 2018 (PS4), May–August 2018 (PS5), September 2018–January 2019 (PS6) and February–May 2019 (PS7). Crop yield data is reported for all seasons. Soil chemical parameters were determined for seasons one to five only, owing to financial constraints. Maize (*Zea mays* L.) was planted in rows and hand weeding was carried out when required.

2.3. SEM

Analysis of the surface of the biochar before and after application to the soil was carried out using the techniques described by Joseph et al. (2010). Imaging was carried out on an FEI NanoSEM 450 and elemental analysis by a Bruker EDS system.

2.4. Shovelomics

The roots of the maize were sampled by using a sharp, flat shovel to initially dig into the soil when maize was harvested for PS2 (July 2017), for all treatments. Following this, the roots were removed from the soil using a cylinder approximately 40 cm in diameter and 25 cm in length, which was placed over the maize plant so that the stem was in the centre of the cylinder. Ten plants were sampled per treatment; two per plot and ten per block ($n = 10$ per treatment; total 50 samples). Representative healthy plants were selected for each plot. The root crowns were cut lengthwise through the middle and carefully cleaned with water, first by soaking them for 3 h followed by rinsing with water for 15 to 30 min. A photograph (resolution 18 megapixel) of the root biomass was taken at constant light conditions on a black background with a HD camera (Canon EOS 60D). The images were analysed using the software REST (Root Estimator for Shovelomics Traits – (Colombi et al., 2015)) which can detect >10 different traits automatically. The images were scaled based on markers present on the picture and the soil surface was set manually on the picture.

To provide a robust measure of the root stock dimensions, REST takes in to consideration only the 95% interquartile width and the 95% interquartile rooting depth. This reduces the impact of single roots protruding the root system to a minimum for these dimension parameters. Within these dimensions, a polygon is placed defining a convex hull that encompasses approximately 90% of the root system. The area of the convex hull is then used as a proxy measure for the root system size and is defined as the area of the convex hull enclosing 90% of root-derived pixels in the image. Traits related to the inner root architecture are more or less independent of root system size. These architecture traits include the fill factor (i.e. the proportion of root-derived pixels within the convex hull), the median gap size (i.e. the size of the holes with visible background within the root system) and the median thickness of measured root clusters. These traits are related to branching density and root numbers.

2.5. Crop and soil data

All maize yield data is expressed as dry grain (ton/ha) following drying overnight at 110 °C. Plant height was measured 2, 4, 6, 8 and 10 weeks after planting. Soil chemical parameters were determined following each planting season up to and including PS5. Soil samples were taken from the top 10 cm of soil from each of the 25 plots and were analysed for the following parameters; $\text{pH}_{\text{H}_2\text{O}}$ (pH in water for 1 g soil

in 2.5 mL water), pH_{KCl} (pH in 1 M KCl), CEC (the sum of cations first extracted by 1 M NH_4OAc (pH 7) and second extracted in 1 M NH_4OAc (pH 7) after reducing the pH to 7.0 via washing with 0.05 M HCl), percent total carbon (TOC, Walkley and Black method), percent elemental nitrogen (TN, Kjeldahl), exchangeable base cations in the CEC extracts and base saturation (via back titration of exchangeable acidity with sodium hydroxide to pH 7), H^+ and Al^{3+} (in 1 M KCl), P_2O_5 and K_2O (in a 25% HCl solution) and available phosphorous (Bray). All methods have previously been described (Alling et al., 2014; Martinsen et al., 2015; Obia et al., 2016; Cornelissen et al., 2018).

2.6. Statistical analysis

Statistical analysis was carried out using R Core Team (2013). All data was checked for normality. Yield and soil property data were non-normally distributed and thus the Mann–Whitney–Wilcoxon rank-sum test was used to test differences between the means of two groups. The full statistical analysis comparing all treatments to each other is presented in the Supporting information, while results for the comparison between biochar and lime and washed biochar and ash are presented in the figures in the main paper and the Supporting information. The shovelomics data was normally distributed and thus ANOVA was used in order to determine if there was a statistically significant effect of treatment.

3. Results and discussion

3.1. Soil and biochar physicochemical properties

Table S1 shows the physicochemical properties of the soil and the biochar prior to amendment. The soil itself was strongly acidic (pH_{KCl} 3.60), with a CEC of 9.7 $\text{cmol}_\text{c}/\text{kg}$, a relatively high exchangeable aluminium content (2.4 $\text{cmol}_\text{c}/\text{kg}$) and a low exchangeable calcium content (0.99 $\text{cmol}_\text{c}/\text{kg}$), resulting in a low Ca/Al molar ratio. These parameters are typical of degraded Indonesian soils and are limiting factors for the growth of crops. The amendment materials (biochar, lime, washed biochar and ash) were all alkaline ($\text{pH}_{\text{H}_2\text{O}}$ ranging between 9.02 and 11.3) and, except for lime, were more nutrient rich than the soil, with different nutrients dominating the different treatments. The biochar and washed biochar contained a mixture of macronutrients, while the lime was dominated by Ca (40.25%) and the ash by K (19.5%). The consequences of these properties are discussed below.

3.2. Effect of treatment on soil properties

Table S2 in the Supporting information shows the properties of the soil following the addition of all amendments for the first five planting seasons (PS), as well as data for the control plot. Fig. 1 shows the change in soil pH_{KCl} , available P (Bray), exchangeable K and Al^{3+} and Fig. S1 shows the change in CEC, Ca, Mg and Ca/Al molar ratio for the different

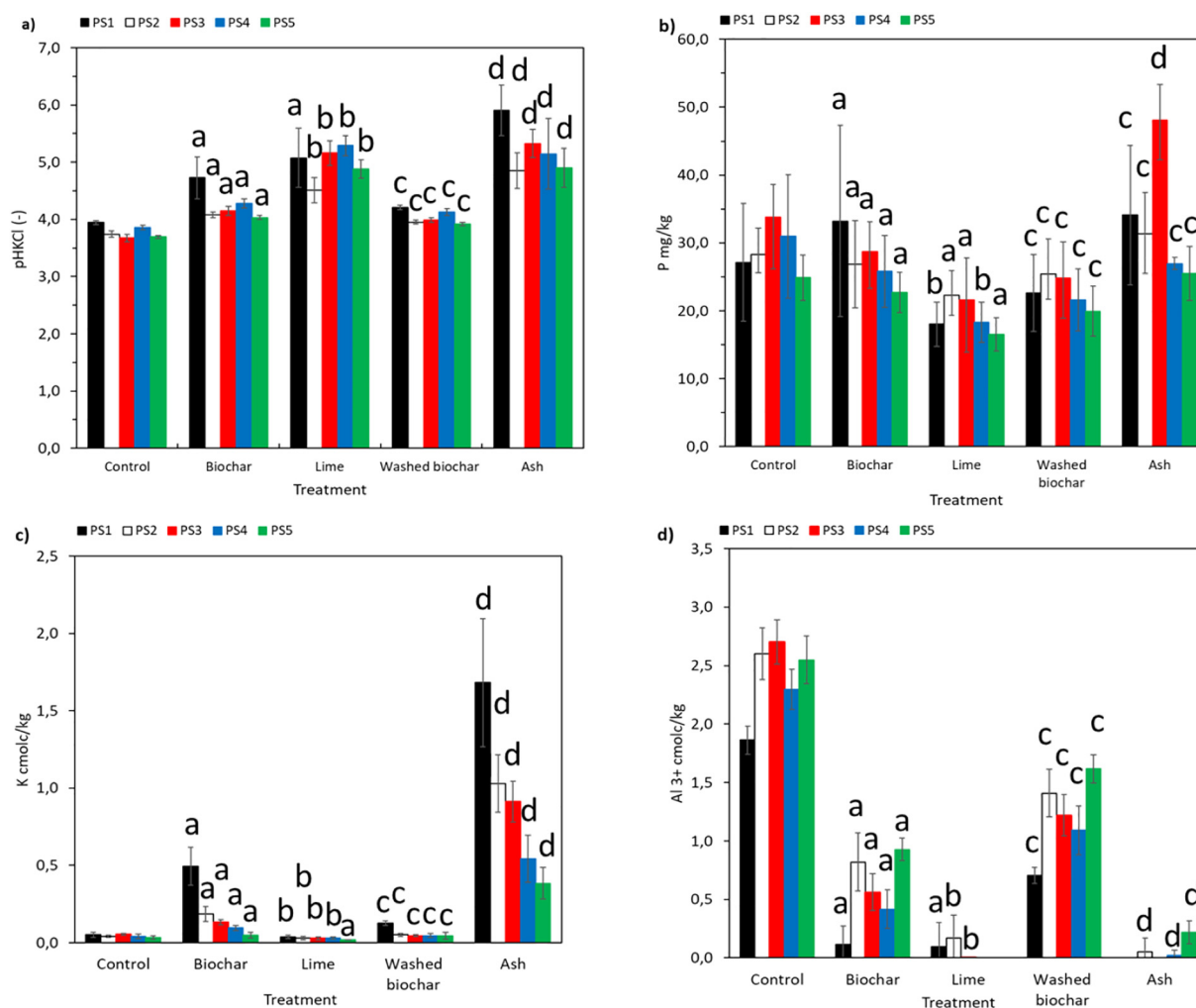


Fig. 1. Soil parameters a) pH_{KCl} , b) available P (mg/kg), c) exchangeable K ($\text{cmol}_\text{c}/\text{kg}$) and d) exchangeable Al^{3+} ($\text{cmol}_\text{c}/\text{kg}$) for all treatments and the first five planting seasons (PS). Statistical results are shown for a comparison between biochar and lime (letters a and b) and for a comparison between washed biochar and ash (letters c and d). For results of the full statistical analysis see the Supporting information.

treatments. Tables S3 shows the results of the full statistical analysis of all soil chemical properties for all seasons compared for all treatments. Soil pH_{KCl} was significantly increased ($p < 0.05$ level) following the addition of all of the amendment materials. All treatments significantly reduced the concentration of Al^{3+} when compared to the control ($p < 0.05$), with the lowest concentrations for the lime and ash treatments (in many cases to below the method detection limit, Fig. 1). A high concentration of Al^{3+} can limit the growth of crops (Dai et al., 2017) and recently a sharp rise in exchangeable Al^{3+} concentration was reported to occur at pH below 4.2 in acidic soils (Gruba and Mulder, 2008). Negative effects on plant roots are expected to occur at lower pH and higher concentrations of Al^{3+} . Both the ash and lime treatments decreased exchangeable Al^{3+} concentration significantly. Previous studies have also demonstrated that biochar can reduce Al^{3+} concentrations to non-toxic levels in acidic soils (Alling et al., 2014), however, although the biochar used here reduced Al^{3+} concentrations significantly ($p < 0.05$), reductions were less pronounced. It is known that alkalinity, associated with carbonates and silicates in biochar can result in Al^{3+} precipitation (Dai et al., 2017), and it is highly likely a similar mechanism is in operation for the ash as this treatment consists of a material enriched in such components. Concurrent with this result, are the Ca/Al molar ratios which can be used to indicate plant stress, where low values represent an increased likelihood of Al saturation. The ratios were the highest for the ash treatment (up to 95), followed by lime (up to 39) supporting the improvement in the soil properties for these treatments. The values for the control treatment were below one for all PS, for biochar were between 2 and 3 (apart from PS) and for washed biochar between 1 and 2.

The change in concentration of the macronutrients phosphorous (P) and potassium (K) that occurred when the amendments were added to the soil, varied depending on the material (Fig. 1). In general, ash resulted in the largest increase ($p < 0.05$ level) in both available K and P concentrations when compared to the other treatments and the control. The soil in the ash treatment had available K concentrations between 0.38 and 1.68 $\text{cmol}_\text{c}/\text{kg}$ depending on the PS, while for all of the other treatments, the concentrations were below 0.15 $\text{cmol}_\text{c}/\text{kg}$. The ashing process, where the organic portion of the biomass material is burned, can result in a material that is enriched in both K and P depending on feedstock and production conditions (Singh et al., 2010). The “ash” contained in the biochar was expected to be present at an equivalent level per kg feedstock material to the “ash” in the ash treatment (i.e. levels of K, Ca, Mg, Na, P and S should be equivalent in both the biochar and the ash treatments), however this did not appear to be the case. The better effect of the ash treatment could be due to a greater availability of nutrients in the ash compared to the biochar. The concentration of exchangeable Ca and Mg were also altered in the treatment plots compared to the control (Fig. S1). Ca and Mg are referred to as secondary plant nutrients, as they are essential to plants, but are required in smaller quantities than N, P and K (Marschner, 2011). For all PS, lime statistically significantly ($p < 0.05$ level) increased the concentration of Ca in the soil compared to the other treatments and the control (apart from when compared to biochar in PS2). For Mg, the ash treatment was able to improve the soil properties to the greatest degree, especially after PS2. Clear effects of time on the soil parameters were not evident, suggesting that the single amendment that took place at the start of the field trial was enough to sustain positive effects over 22 months.

3.3. Effect of amendment on crop yield

Fig. 2 and Table S4 show the maize dry grain yield (ton/ha) for all treatments over seven PS. The control plot receiving just NPK fertilizer and urea had a low yield (1.22 ton/ha in PS1 and 0.4 ton/ha in PS2, 0 ton/ha in PS3 to 7). It is possible that there was a small yield in the first two PS as the field had been fallow for several years prior to starting the experiment. This reiterates the low productivity of these degraded

acidic Indonesian soils and highlights the need for treatments that can improve their fertility and productivity. A recent meta-analysis investigating the effect of the amendment of biochar alone or biochar in combination with inorganic fertilizer, showed that on average, a 26% increase in yield was achieved for the inorganic fertilizer alone treatment, and an increase of 48% was observed for the biochar and inorganic fertilizer combined treatment, when compared to the control non-amended treatment (Ye et al., 2019). All treatments used in the field experiment increased the yield by an average of; seven times for the biochar treatment, five times for the lime treatment, five times for the washed treatment and eight times for the ash treatment, over the seven PS.

The treatment that resulted in the highest yield in PS1 and PS2 was biochar, however the yield was not statistically significantly different ($p > 0.05$) from any of the other treatments (see Table S5). From PS3 and onwards, ash resulted in the highest yield (statistically significant, $p < 0.05$), lending support to the notion that macronutrients are more available in this treatment than in the biochar treatment. In the majority of cases, the washed biochar was the treatment (apart from the control), that resulted in the lowest yield, and in general the following order of treatment effectiveness was; ash>biochar>lime>washed biochar>control. Ash is known to contain the biogenic elements (apart from N) necessary for the proper growth of plants and can supplement deficiencies of micro and macro elements in soils (Saletnik et al., 2018). These crop yield results support those observed for the soil, as it was ash that provided the greatest concentration of P, K, Mg, the largest pH and the largest Ca/Al ratio, all of which are conducive to better plant growth.

The literature related to previous long-term maize field trials in which biochar, lime, washed biochar and ash have been used as amendment materials is limited (Kahl et al., 1996; Moragues-Saitua et al., 2017; Reed et al., 2017; Richard et al., 2017; Saletnik et al., 2018). In a Polish study, both biochar and ash were added to soil and the effect on *Miscanthus x giganteus* was monitored. Depending on treatment and dose, there was an increase in yield of between 8 and 68% and the positive effects were ascribed to the increase in pH from 5.0 to 6.3 (Saletnik et al., 2018). Kahl et al. (1996) carried out a two-year field trial in which ash produced from birch, beech, red spruce and balsam fir was added to an acidic sandy soil. An increase in exchangeable nutrient cations and pH (from 3.5 to approximately 6) as well as a decrease in available Al concentration (up to 70%) was reported for ash doses up to 38 t/ha. In a study to directly compare biochar and ash, neither treatment resulted in a significant effect on the growth performance of grass, despite an increase in soil pH (original soil pH without amendment was 6). The authors speculated that the soil quality was good enough for optimal grass growth prior to the addition of other materials (Reed et al., 2017). The varying results in these studies highlights the fact that effects on soil properties and crop yield are often not straightforward to predict and can be a combination of many factors.

In a field study carried out for six years in China, biochar was added to an acidic soil at seven doses and rapeseed was grown. The positive effects on yield were significant for all doses over 10 t/ha when compared to the control unamended plot and this was related to an improvement in the soil hydraulic and acidity properties. The soil used was an upland red soil with a pH of 4.24, which was increased by 0.53 units following amendment (Jin et al., 2019). It is possible that in this field study, the use of the washed biochar when compared to the control treatment, provides an indication of any physical effects of treatment. The washed biochar may contribute positively to soil moisture retention; however, this would need to be investigated more fully before firm conclusions can be drawn.

A previous study that investigated biochar and lime treatments showed that both significantly influenced soil pH, exchangeable Al and available nutrients. However, in that study, while biochar demonstrated a positive effect on rice bean yield, the addition of lime resulted in a decrease of available P which is non-conducive for plant growth (Yao et al., 2019). Mensah and Frimpong reported a positive effect (up

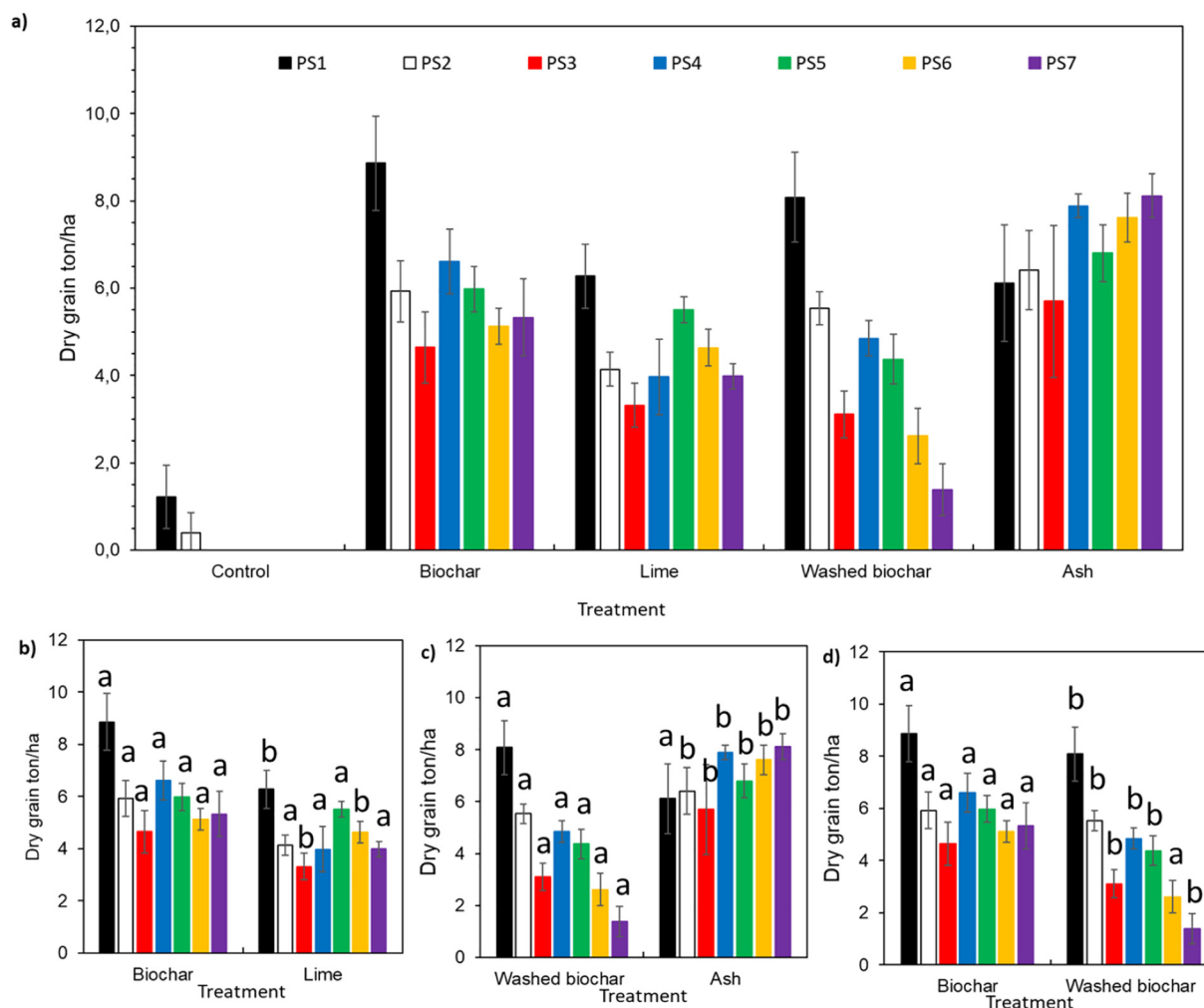


Fig. 2. Maize yield (dry grain in ton/ha) for 7 planting seasons (PS) for all treatments: control, biochar, lime, washed biochar and ash. Data is shown in a) for all treatments without statistical results, b) for biochar compared to lime in addition to statistical differences, c) washed biochar compared to ash and d) biochar compared to washed biochar, in addition to statistical differences. Statistically significant differences are at the 0.05 level. Different letters indicate a difference between the treatments biochar and lime, washed biochar and ash, and biochar and washed biochar (letters a and b). For complete statistical analysis results see the Supporting information.

to 300% increase in dry matter) on the growth of maize when 2% biochar was added to an acidic soil in Ghana (Mensah and Frimpong, 2018). The authors reported an increase in soil pH from 4.6 to 5.7, a reduction in available Al concentration and an increase in the concentration of both micro- and macronutrients.

This field trial was carried out for seven planting seasons (22 months) and different time trends are observed for different treatments. For biochar and lime, a decrease in effect is observed from PS1 to PS2, however this is then maintained from PS3 to the end of the trial. For the washed biochar treatment, the effect on yield declines over all PS. For the ash treatment, the effect is sustained over all PS. Previous studies have also shown that the effect of biochar amendment can decrease with time (Cornelissen et al., 2018) as the positive effects the biochar addition had on the soil are reduced. Carrying out this field trial over 22 months supports the notion that the benefits the ash supplies to the soil are sustained over this period, while the benefits from the other treatments are more short lived (Reed et al., 2017).

3.4. Liming effect

Prior to beginning the field trial, it was hypothesized that one way in which the treatments could increase the yield of maize was via a liming effect through which soil pH increases, while exchangeable Al decreases. A comparison of the lime and biochar treatments provides

support of this hypothesis. This effect is observed when the pH of the soil increases to a level at which the negative effects of plant available Al and deficiencies of available P are overcome (Jeffery et al., 2011; Yao et al., 2019). A sentinel study by Yamato et al. (2006) provided direct evidence of an increase in pH and decrease in available Al^{3+} concentration when biochar was added to an acidic Indonesian soil. Despite attempts to obtain the same pH for both the biochar and the lime treatments, the pH in the lime treatments was statistically significantly higher ($p < 0.05$) than the biochar. The pH for biochar treatments ranged between 4.03 and 4.73 over the five PS, while for lime the pH was between 4.51 and 5.29. Consequently, the exchangeable Al^{3+} concentration was statistically significantly lower for the lime compared to the biochar treatment for all PS other than PS1 ($p < 0.05$), and the Ca/Al was significantly higher for the lime treatment ($p < 0.05$) (see Fig. 1 and S1). Biochar is able to reduce the concentration of available Al^{3+} through a complexation of Al with its surface oxygenated functional groups and a crystallization of gibbsite (Berek and Hue, 2016).

Despite the fact that the lime treatment resulted in the highest pH and the lowest Al^{3+} concentrations in soil, it was the biochar treatment that produced the highest maize yield when biochar was compared to lime (significantly for 3 PS, see Fig. 2b). Importantly, this results shows that while there is a liming effect at play, there are additional properties of the biochar that further improve the soil and result in a more conducive growth environment. Biochar can be made for as little

as 100 US\$/t by these smallholder farmers, especially when using clean, fast and free-of-charge flame curtain kilns (Cornelissen et al., 2018), compared to a dolomite price of 250 to 500 US\$/t, as dolomite is used to lime in Indonesia. The comparison between the biochar, washed biochar and ash treatments also supports the conclusion that there are other factors involved in the resultant effects. Washing the biochar significantly reduced the pH and increased the plant available Al^{3+} over all PS ($p < 0.05$), compared to the un-washed biochar. These factors in turn led to a lower yield for the washed biochar treatment (significant for all PS, Table S5). The ash treatment had a higher pH and a lower Al^{3+} concentration compared to the biochar (significant in all cases for all PS, except one case ($p < 0.05$)) and a greater yield (significant for all PS, $p < 0.05$).

3.5. Nutrient addition effect

The second hypothesis postulated prior to beginning field trials was that effects on yield could also be explained by a nutrient addition effect, whereby the different treatments result in an increase in essential soil and plant macro and micronutrients which in turn improve maize yield. Results from the field trial support the fact that there is a nutrient addition effect at play. The washed biochar and ash treatments are those treatments that are most dissimilar to each other in this respect: the water washing of the biochar removes a proportion of the ash fraction which contains important macro and micro nutrients, which is all that remains in the ash treatment (Richard et al., 2017). The maize yield for the ash treatment is statistically significantly higher ($p < 0.05$) than the washed biochar treatment (Fig. 2c). It is the ash treatment that contained the greatest concentrations of some of the most important nutrients; P, K, Ca and Mg, and concentrations were statistically significantly higher ($p < 0.05$) in the ash treatment than the washed biochar treatment for all seasons and all nutrients. Following the previous discussion it seems plausible that these nutrients are more available than for the biochar treatment and hence result in the greater effect.

The base saturation (shown in Table S2) showed a similar trend, and these combined provide direct evidence that the ash treatment is providing a nutrient addition effect in addition to its pH effect, resulting in the highest maize yield. When comparing the biochar to the washed biochar (Fig. 2d), it appears that the nutrients remaining after washing are enough to sustain crop growth in the short term but not in the long term. Maize yield is similar for PS1 for these treatments, but higher for biochar than washed biochar for the subsequent seasons.

The most striking nutrient result was observed for exchangeable K where concentrations in the soil for the ash treatment peaked at 1.68 $\text{cmol}_\text{c}/\text{kg}$ for PS1 compared to 0.13 $\text{cmol}_\text{c}/\text{kg}$ for the washed biochar, 0.49 $\text{cmol}_\text{c}/\text{kg}$ for the biochar and 0.04 $\text{cmol}_\text{c}/\text{kg}$ for the lime. This result supports the notion that the nutrients provided by the ash treatment are more available than for the other treatments. The observation that yield was highest for the ash treatment is thus probably explained by a combination of an increase in pH, resulting in a decrease in plant available Al^{3+} , as well as a great increase in exchangeable K. Haefele et al. (2011) carried out a field study in Thailand and the Philippines and concluded that the positive effect on yield over a four year period was due to an increase in K concentration and water retention. In Lampung, the high level of precipitation suggest that crop growth caused by water limitations are not likely a problem. In a previous field trial carried out in the same soil as used here and for 24 months (Cornelissen et al., 2018) reported a fading effect of the amendment of biochar at two different doses (5 and 15 t/ha) produced from two different materials (cacao shell and rice husk) as the acidity alleviation effect declined over time. This was postulated to be due to a leaching of alkaline ashes which is known to occur in humid tropical climates that have prolonged or large rainfall events (Major et al., 2010a; Glaser, 2012). The results of this field trial suggest that such an effect had not started to occur after the 22 months this trial was carried out for and could possibly be due to the coincidence of the field trial with an El Nino event.

3.6. Shovelomics

Apart from changes in soil quality, changes to the plants themselves can also explain crop effects of biochar. Shovelomics can be used in order to produce high throughput data from field studies (Trachsel et al., 2011) in order to assess effects on root characteristics following soil treatment as root system architecture is one of the major factors that determines biomass productivity. All treatments resulted in the alteration of the traits identified by shovelomics, and in all cases the differences were significant apart from the fill factor. The rooting depths were approximately 25% higher, the width approximately 50% broader and the area approximately 75% larger than the control for all treatments. Fig. S2 in the Supporting information provides photographs of the effects on the root system for the different treatments. In concurrence with the maize yield results, it is the ash treatment that had the strongest effect on root angle opening (75.90 ± 21.23), area (50.47 ± 12.37) and stem diameter (1.09 ± 0.39). There were very few differences between the other treatments. These data show that all treatments improved the nutrient status of the soil, when compared to the control treatment and it follows that all root systems benefit from these changes. As has previously been observed, the root biomass and traits do not directly mirror the yield dynamics (Abiven et al., 2015; Hirte et al., 2018), for the treatments apart from ash. In this field trial it appears that the rooting systems reached their potential for all treatments, and that the improvements in yield are more related to a better resource use efficiency, than a better exploration of the soil. The slightly better performance of the ash treatment is likely a result of the greater availability of the nutrients.

3.7. Microscopic analysis of the biochar

High magnification images and EDS spectra (to provide elemental information) of the biochar prior to amending it to the soil reveal the presence of mineral clusters, possibly carbonates and/or oxides of Mg, Ca and K, attached to the surface of the biochar (Fig. S3, point a). In addition, the biochar pores are seen to contain more complex mineral clusters rich in Mg, Al, Si, K, P, Fe and Ca, bonded with organic compounds (Fig. S3, point b). Following amendment to the soil, an organomineral layer with a high concentration of Al and Si compounds (and possibly clay minerals) was observed to have formed on the biochar surface and in the biochar pores (Fig. S4, point a). Interestingly, mycorrhizal fungi can be seen to have grown into this organomineral layer, possibly penetrating biochar pores as has been reported previously (Fig. S4, point b) (Blackwell et al., 2015; Joseph et al., 2015). The corresponding EDS spectra suggest that the mycorrhizal fungi have populated an area relatively rich in Mg, Fe, K and Ca. These observations may in part (along with the liming and nutrient addition effect), explain the good performance of the biochar amendment.

4. Environmental sustainability

The results from this field trial showed a very positive effect of the ash treatment on the performance of maize. Ash contained more available nutrient cations than biochar made from the same amount of feedstock and the effect of the ash treatment was also more long-lived than that of biochar. In this degraded acidic Ultisol there are many environmental, practical and financial factors that must be considered when small scale farmers decide whether, and in what way, they would like to treat their soils. Despite the positive results, completely burning crop residues to produce ash is not advocated owing to both the negative environmental effects this causes, and the positive environmental effects that are lost. Air pollution and the release of green-house gases increases with the production of ash. In addition, the benefit of carbon sequestration (Woolf et al.,

2010) that results when biochar is made is not realized when ashes are produced. Crop residues themselves represent a potentially important source of soil organic matter, important for soil fertility and soil moisture regulation. Therefore, small scale farmers in Indonesia with degraded acidic Ultisols are advised to produce biochar from their waste agricultural material as this not only provides an increase in crop productivity, but also sequesters carbon resulting in the best overall environmental benefit.

CRediT authorship contribution statement

Sarah E. Hale: Funding acquisition, Conceptualization, Methodology, Writing - original draft. **Neneng L. Nurida:** Conceptualization, Methodology, Writing - review & editing. **Jubaedah:** Methodology, Writing - review & editing. **Jan Mulder:** Conceptualization, Methodology, Writing - review & editing. **Erlend Sørmo:** Methodology, Writing - review & editing. **Ludovica Silvani:** Methodology, Writing - review & editing. **Samuel Abiven:** Methodology, Writing - review & editing. **Stephen Joseph:** Methodology, Writing - review & editing. **Sarasadat Taherymoosavi:** Methodology, Writing - review & editing. **Gerard Cornelissen:** Conceptualization, Methodology, Writing - review & editing.

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Declaration of competing interest

The authors declare no conflicts of interest.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2020.137455>.

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